

Efficient and Sustainable Remediation of Refinery Wastewater Using Electrocoagulation and Advanced Oxidation Techniques

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ABSTRACT

Effluent wastewater from industrial processes needs to be properly treated before being discharged into the environment. Conventional procedures for handling this wastewater can be problematic due to the presence of toxic elements, time constraints, and complexity. However, a new electrochemical procedure has been developed as an effective method for remediation. In a recent study, refinery wastewater was successfully treated using an electrochemical technique combined with ultrasonic irradiation and photocatalysis. The study found that electrocoagulation, which uses cheap and recyclable metal electrodes, was a simple, efficient, practical, and cost-effective way to handle refinery wastewater. Various parameters were investigated, including electrode metals, operating time, applied voltage, pH, inter-electrode gap, and temperature. The aim was to determine the optimal configuration for pollutant removal. The study also focused on the synergistic effects of combining electrocoagulation and photocatalysis to improve the efficiency of contaminant removal in oily wastewater. By integrating these two treatment technologies, the researchers aimed to enhance pollutant removal rates, energy efficiency, and overall system performance. The research provided valuable insights into the feasibility, optimization parameters, and applicability of the electrocoagulation-photocatalysis process for remediating organic contaminants in oily wastewater industrial effluents. The results showed that electrocoagulation, especially when combined with ultrasonic irradiation and TiO₂ photocatalysis, was highly effective in pollutant removal within a short timeframe. These findings support the implementation of this procedure for remediating most industrial wastewater. In conclusion, the study contributes to the development of more effective and sustainable water treatment strategies. The electrocoagulation-photocatalysis process shows promise in addressing the remediation of organic contaminants in oily wastewater from industrial processes.

Keywords: electrocoagulation, photocatalysis, ultrasonic, wastewater, refinery, oily wastewater

INTRODUCTION

The growing shortage of fresh water is a worldwide concern. A substantial reduction in the amount of obtainable fresh water (in both quality and quantity) is exacerbating fears about how this will influence human well-being, ecosystems, and

the world economy (Al-Jadir et al., 2022). Thus, many countries and organizations have attempted to solve this problem by reducing contamination, eliminating dumping, decreasing the number of unsafe materials that are released, reducing the quantity of untreated wastewater, and greatly increasing reprocessing and safe recycling.

It is apparent that evolving manufacturing and urban activities are key factors in world economic improvement. However, ignoring the ecological consequences of improper waste remediation and dumping will eventually negatively affect the environment greatly, even influencing climate alteration in the long term.

For example, Iraq, one of the largest international oil manufacturers, generates about 20 million tons of crude oil annually; with every ton of oil obtained, a high volume of water is consumed for extraction, with about 50% of the water released into the environment [UNEP, 2007]. Furthermore, the discharged water is highly polluted by inorganic and organic elements. Some nontoxic nutrients can actively facilitate the production of algae and eutrophication, where traditional procedures may be unsuccessful to remediate this ecologically. Likewise, most wastewater effluents from many industries are released into rivers after performing conventional treatments in special plants [Sun et al., 2018; Al-Rubaiey, 2022]. Typical conventional treatment procedures conducted before discharging water into rivers may involve many standard processes (i.e., physicochemical, biological, advanced oxidation) and other evolving technologies. The nature of these treatments differs based on the type of wastewater, including its impurities. Advanced oxidation processes need effective oxidants for the remediation to be both safe and cost-effective. Alternatively, biological processes require firmly controlled conditions with a long operating time, often resulting in undesirable by-products. Chemical processes require adding various substances that not only improve the practice charge but also create downstream problems, with a greater hazard of further impurities. For example, membrane filtration alone cannot proficiently handle wastewater without being combined with other pretreatment procedures. Otherwise, the systems become unproductive over time due to pore hindering and reduced flux [Sathya et al., 2022]. Consequently, this wastewater remediation study has been incorporated with new electrochemical systems, known as electrocoagulation (EC).

In 1889, electricity was first adopted in water remediation in the UK, while the EC procedure was discovered in the U.S. in 1909 [Vymazal, 2022]. Although the EC was practiced in the U.S. in industrialized softwater plants in 1946, it was unsuccessful in achieving global acceptance for broader uses and was restricted by the cost of

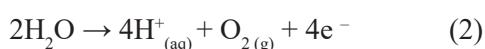
electricity and the enormous capital investment required [Chen and Hung, 2007]. Nevertheless, continuous development in electrochemical studies supported the use of the EC in the remediation of wastewater [Sathya et al., 2022]. Among all electrochemical systems, the EC stands out as the optimal method because it is eco-friendly and a sustainable substitute to handle wastewater due to its simplicity, small footprint, and capability for dealing with a large amount of fluid waste with no broad chemical aspects. Additionally, the adaptability of the procedure and its arrangement allows the EC process to handle an extensive range of waste throughout industries with diverse types of contaminants. Many electrochemical investigations have been performed globally to remediate numerous forms of liquid waste and have accomplished encouraging products [Chen and Hung, 2007]. Some studies have combined the EC process with other procedures, creating advanced oxidation systems that have yielded cleaner and safer discharges [Muttaqin et al., 2022; Mengistu et al., 2022; Magnisali et al., 2022; Rookesh et al., 2022; Asfaha et al., 2022; Rakhmania et al., 2022; Tahreen et al., 2020; Bagastyo et al., 2022]. The current paper reports recent progress in the evolution of the EC and its emerging hybrid processes in treating industrial wastewater.

ELECTROCOAGULATION

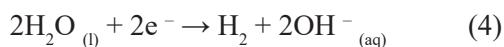
The EC procedure is part of an electrochemical technique that destabilizes the charges of impurities using an electric current to create electrode disintegration and catch pollutants in flocs that can later be easily detached from the solution. The ions produced from the metal dissociation and the following hydrolysis both act as coagulators that serve to reduce the number of charged contaminants and push these impurities to meet the shaped floc that collects them. Due to their polar momentum and reaction capacity, the produced hydroxyl ions produce intermediate complexes combined with the waste product, thus causing larger floc creation. An EC cell arrangement is comprised of a power source and Al/Al metals submerged in wastewater within an isolated container for remediation [Muttaqin et al., 2022].

The key parameters that may influence the EC procedure are the distance between the inter-electrodes (i.e., inter-electrode gap), electrode type, applied voltage, conductivity, initial pH,

and run time. Therefore, these operating variables were varied in each EC investigation to obtain optimum conditions. Typically, metals (e.g., Al/Al) are adopted as EC electrodes because they are cheap, abundant, and cause extraordinary pollutants removal efficacy. These electrodes discharge Al/Al hydroxide ions into the solution due to the voltage passed through them, providing a great affinity to react with the water impurities. The following equations summarize the initial reactions in this process, where M represents the metal electrode, and n represents the charge number of the ion [Mengistu et al., 2022]. The starting reactions at the anode are symbolized by the following:



Whereas the starting reactions at the cathode are symbolized below:



As these reactions advance, the freeing of the produced ions from the electrodes' detachment can be predicted with Faraday's law [Tahreen et al., 2020]. The theoretical depletion rate of sacrificial electrodes evaluated using this law estimates the probable conduct of the EC. Additionally, the beneficial properties of EC greatly exceed those of conventional chemical coagulation (CCC). This simple and easy EC procedure eliminates the moving parts and repairs needed by other methods. Compared to CCC and many other wastewater treatments, EC produces a radical change in the chemical oxygen demand (COD) removal efficiency [Aswathy et al., 2016]. And compared to CCC, the produced sludge from EC is acid resistant, settles easily, and has high stability, a larger floc size, and less affinity for water that could be separated with filtration. Moreover, using electricity as a motivating force for Al/Al coagulant creation reduces the need to add chemicals. Recently, outstanding EC process-based wastewater remediation has been obtained using solar energy, which is beneficial as electricity is a cost-restrictive factor [Sharma et al., 2011; Naje et al., 2016]. By integrating renewable solar energy as the power supply, EC can add to both the commercial and ecological sustainability of wastewater treatment technology.

Concluding our exploration of the current state of relevant wastewater treatment techniques,

it becomes evident that there exists a critical gap in our understanding of organic contaminants removal. The limitations of existing methodologies and the persistent challenges in wastewater treatment underscore the need for innovative and integrated approaches to address these complexities. Therefore, this study aims to investigate the synergistic effects of electrocoagulation combined with photocatalysis with a focus on enhancing the efficiency and sustainability of effluent wastewater. By filling this gap and elucidating the potential benefits of the proposed approach, our research contributes to the advancement of knowledge in water remediation and offers practical insights for industrial application.

OILY WASTEWATER PROBLEM

Nowadays, many industries generate a great quantity of oily wastewater, which causes various adverse impacts on the surrounding environment and sanitary conditions. Many countries are setting regulatory limits on the maximum oil concentration in oily wastewater discharge to be within the 5–100 mg/L range. To clean oily wastewater, many methods are forecasted and classified into chemical, physical, mechanical and biological approaches. Gravity separation (GS) and dissolved air flotation (DAF) can be classified as physical methods to clean oily wastewater. Currently, GS is being used as the first stage separation process for dispersed and floating oil, and it is not applicable for the separation of emulsified oil. In the 1990s, many studies had been conducted to evaluate the effectiveness of gravity separators in oil spills, and these studies focus on the efficiency of the separators after the weathering effect on the oil spills, mathematical modeling of the mechanism in the separators and the design of separators to warrant ease of operation for variable fluids and operating conditions. GS is a very simple system, but it has many disadvantages like limited separation capacity, requires a large area for setup and complex management and operation. The principle of DAF is to introduce air under pressure at the bottom of an open basin, and as the air bubbles rise to the top of the basin, it will bring along pollutants [Abuhasel et al., 2021; Sathya et al., 2022].

Despite the application of all the technologies used, oily wastewater treatment technology is still energy-intensive, unstable, needs high operational and installation costs and does not produce

the expected yield. The application of advanced techniques, nanotechnology and integration of the system is required for efficient and cost-effective oily wastewater treatment. The information presented in Figure 1 illustrates the range of pollutant concentrations observed when applying the Electrocogulation technique, in contrast to alternative methods. It showcases both the minimum and maximum levels of pollutants examined.

MATERIALS AND METHODS

Detailed explanations of this investigational work are stated elsewhere [Al-Rubaiey and Al-abrazanjy, 2017; and Al-Rubaiey et al., 2022]. TiO₂ was obtained from US Research Nanomaterials Inc. (U.S.), with a 25-nm particle size. Other chemicals were purchased from Merck (Germany). All chemicals were used without further handling. A photograph of the investigational setup is shown in Figure 2.

The practical work was performed in a bench-scale reactor with Al/Al electrodes (containing 17 holes of Ø6 mm, distributed throughout) connected in a monopolar mode to a DC power source. The φ45 × 2 mm, plates had a total effective area of 11.0 cm² and were located horizontally in the reactor at an optimum distance of 1 cm apart. The metal electrodes were coupled to a DC standard power supply (0–20 V) output voltage, connected with a multimeter. Prior to each experiment, the electrodes were slightly sandpapered, then washed and rinsed with distilled water to remove any passive layers from their surfaces. A lab magnetic mixer (BOECO MSH-300N, Germany) was

employed to mix the electrolyte solution during the remediation. Hanna multimeter (HI-9828, Romania) multi-sensor probes were utilized to read variations in the pH, total dissolved solids (TDS), and temperature.

Water samples from the solution were collected intermittently and examined with a standard turbidity analysis technique and recorded using a Hanna system water analysis turbidity meter (HI93703, Romania). A commercially available ultrasonic bath (Silver Crest, 46 kHz, 50 W, China) was operated to examine the impact of ultrasonic waves on the EC system. This study only examined the physical characteristics of oily wastewater obtained from the Al-Dora refinery (Baghdad, Iraq). Its initial turbidity concentration, pH, and temperature were 500 FTU, 6, and 35°C, respectively. For each experiment, 1 L of wastewater was tested. The pH of the wastewater was not adjusted so as to be able to evaluate the efficiency of the process without adding any chemicals. The turbidity removal efficiency, RE%, was evaluated using Eq. 5 (Jabbar et al., 2023; Alardhi et al., 2023):

$$RE(\%) = \frac{(C_o - C_t)}{C_o} \times 100 \quad (5)$$

where: C_o – the turbidity at $t = 0$, C_t – the turbidity at an exact time.

For photocatalysis runs, all the work was conducted in a batch photoreactor [Al-Rubaiey et al., 2022]. The solution was mixed using a magnetic stirring procedure. The radiation source was a UV lamp (30 W, UV-C, $\lambda_{max} = 254$ nm, manufactured by Philips, The Netherlands) which was situated inside the Pyrex® reactor. During the UV irradiation, the reactor was mounted on a magnetic

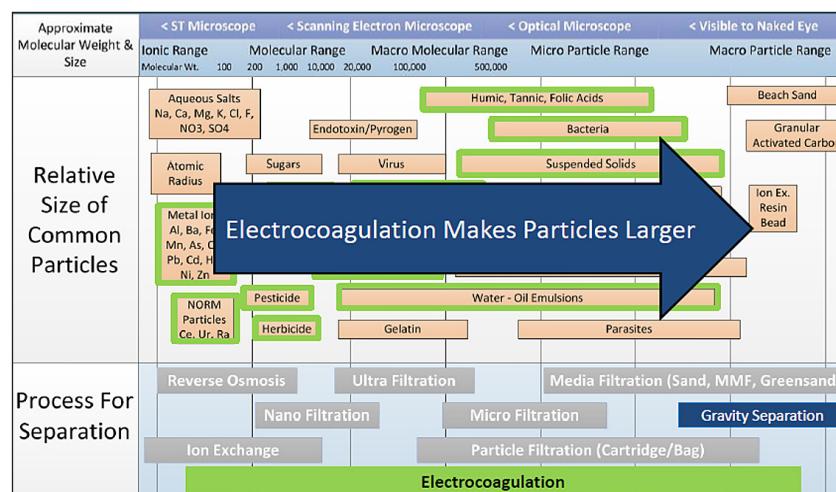


Fig. 1. The range of pollutant concentrations observed when applying the electrocoagulation technique



Figure 2. EC experimental setup

stirrer to maintain the suspension's homogeneity; and at 15-min reaction intervals, 10 ml of the solution sample was collected and analyzed. TiO_2 as a nanoparticle was supplied to the liquid waste to stimulate reactions. At that time, the flat-plate electrodes (Al) were immersed in the solution and connected to the power supply. In each run, 800 ml of the water was used at 15 min course intervals. Then the power supply and UV lamp were turned off to allow precipitation for several minutes (≈ 5 min). At that point, the resulting water was tested.

The COD tests of the solution were measured using a standard method, the open reflux method, 5220B [Ngala et al., 2019], for the inspection of sampled waste liquid. The current density and pH was controlled during electrocoagulation process. The process is done at pH average value of 8, current density of 45 mA/cm². The current density is a crucial parameter influencing the efficiency of electrochemical processes. To address this, we have calculated the current density based on the electrode's circular shape with a diameter of 4.5 cm and an average current of 500 mA. The resulting current density is approximately 45 mA/cm².

RESULTS AND DISCUSSION

Effect of applied voltage

Figure 3 shows the outcome of using various voltages on the removal efficiency (RE%) of oily wastewater with an optimum electrode distance of 10 mm, at 308°K. The results demonstrated

that the RE% of the process was directly proportional to the values of the used voltage. This parameter is essential in the EC procedure as it controls the dosage rate of the coagulant, the production rate of the bubble, the bubble size, and the development of the flocs. This parameter regulates the electron discharge rate as a result of the detachment of ions from the anode. In fact, the anode disintegration was directly proportional to the applied voltage. However, the values of this voltage fluctuated broadly for the diverse types of wastewater. The variations were primarily due to the discrepancy in the ionic contact caused by the features of the contaminants found in the waste. Even though this is greatly related to the Al/Al detachment and discharge of ions into the electrolyte solution, any extra voltage may also negatively affect the efficacy of EC by allowing further reactions to occur; and the extra levels of coagulants may provide charge setback of the colloids. This behavior would shorten the electrodes' lifetime and worsen the RE% of the EC and the lifetime of resources (e.g., electrodes, the power supply, etc.). Thus, this is a significant feature that necessitates ideal conditions to perform the EC process for the anticipated remediation. High voltage may lead to a great drop in the ohmic value between the Al/Al electrodes, causing higher operating costs. However, by lessening the applied voltage, energy consumption costs are manageable, but this also compromises the effectiveness of the reaction time. Consequently, a better solution to obtain the same EC RE% could occur by adapting other crucial factors of EC (i.e., increasing

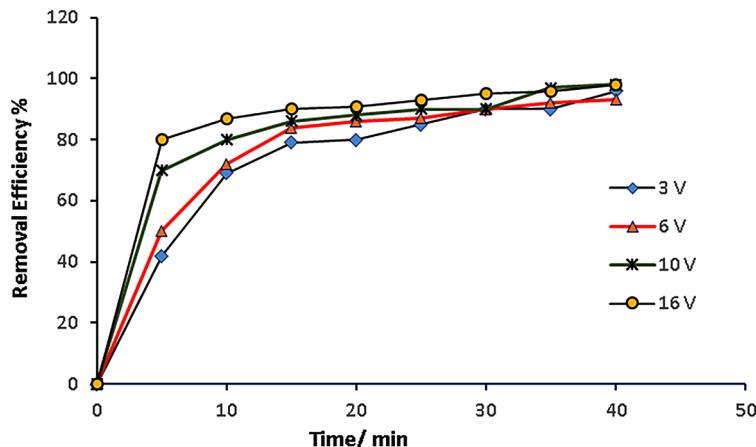


Figure 3. Effect of the applied voltage on the RE% of the EC wastewater treatment

conductivity, reducing the inter-electrode distance, and increasing the electrodes' surface area) [Ngala et al., 2019; Setyawati et al., 2021; Changmai et al., 2019; Chen et al., 2018; Demirbas and Koby, 2017; Um and Chang, 2017].

Effect of the electrode material

The materials employed in manufacturing the EC electrodes hugely influence the system's performance, RE%, and cost. These materials also govern the type of starting reactions that can be initiated during the electrochemical process [Al-Rubaiey et al., 2020; Al-Rubaiey and Albrazanjy, 2019; Al-Rubaiey and Albrazanjy, 2018a]. The RE% is effectively arbitrated by the metal disintegration rate, the impurity RE%, and the type of Al/Al coagulant chosen. These features are directly linked to the discharge of the Al/Al ion hydroxyl in the waste liquid. Since these materials have a high degree of electrical double-layer density, Al/

Al floc coagulants with a greater valency are selected because of their ability to enhance the contaminant coagulation phase. Due to their many beneficial properties (e.g., their ability to produce active coagulation, cost-effectiveness, accessibility, dependability, and nonhazardous features), Al/Al metals have been picked universally by researchers for EC systems, where they have effective RE% of treated impurities. Although both Fe and Al metals have comparable levels of reactivity, variations in their ion dissociation resulted in different electrochemical consequences. This is shown clearly in Figure 4, which illustrates comparable trends for both metals. However, there are conflicting views concerning the most fitting electrode arrangement for particular wastewater remediation [Linares-Hernández et al., 2009]. Variations in applicable parameters (for instance, operating time, solution features, and pH) may jointly influence the EC's efficacy [Chafi et al., 2011]. In addition, many studies have found that

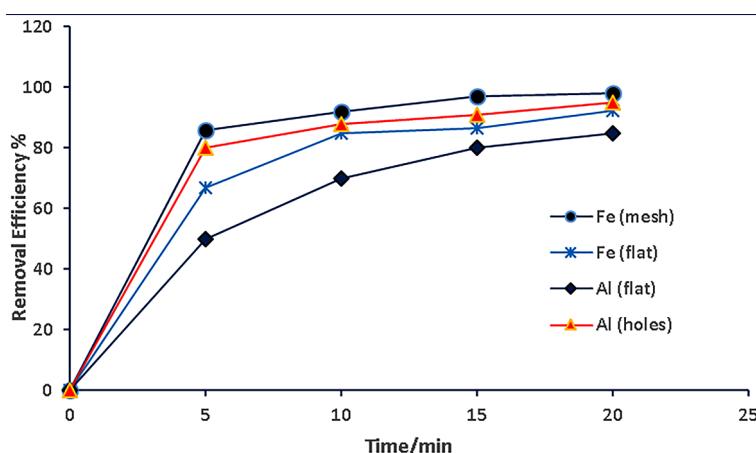


Figure 4. Effect of the electrode material on the RE% of the EC wastewater treatment

both Fe and Al have resulted in the same RE% values, with Fe generally being more efficacious than Al [Naje et al., 2017]. Similarly, in some investigations, Fe produced the maximum decolorization with lower energy consumption and cost than Al [Linares-Hernández et al., 2009]. Therefore, it is difficult to decide which EC arrangement would provide the best result because Fe and Al possess particular, distinctive characteristics in relation to specific circumstances. Additional parameters, for example, the optimum operating time and initial pH, should be tested carefully as they differ based on the electrodes being used and depending on the wastewater chemistry. Nevertheless, some scientists have established that an Al/Al arrangement is the preferred setup for maximum EC RE% [Linares-Hernández et al., 2009; Naje et al., 2017]. Therefore, the latter combination was adopted in most of our experimental configurations.

Effect of the Inter-electrode distance

Proper-sized gaps between the Al/Al metals are required in the electrochemical procedure because these gaps govern the static field initiated among the electrodes. This field is optimal when the inter-distance is reduced to the lowest values [Kobya et al., 2015]. Hence, the formed Al/Al hydroxides are necessary to shape the flocs and to sustain the coagulation, which degrades due to the strong collisions from the high electrostatic interaction. Thus, the value of the RE% was lowest with the smallest inter-electrode gap. However, a larger inter-electrode gap may interfere with the development of the hydroxide flocs that have resulted from the lower electrostatic forces.

Gapping outside of the ideal value reduces the RE%, requiring greater energy to overwhelm the slower interaction of the discharged ions passing between the two electrodes. Accordingly, it is paramount to operate EC with an optimum gap length. Various distances (averaging 10 mm) have been employed to treat many types of wastewater [Kobya et al., 2015; Elazzouzi et al., 2017; Zeboudji et al., 2013; Khaled et al., 2019], and this distance has been supported by the current study.

The results shown in Figure 5 present the effect of EC efficiency with modifications in the inter-electrode gaps (5–20 mm). From Figure 5, it can be seen that as the electrode gap decreased, the pollutants' RE% directly increased; as mentioned before, this may be due to the reduction in the wastewater resistance with the lowest inter-distance between the produced Al/Al hydroxyl ions. Hence, at a smaller amount of applied voltage, a greater RE% was attained. However, as with all crucial features, the most fitting gaps also rely on the type of waste solution and the reactor arrangement. A larger gap between the Al/Al metals can be overcome by a higher amount of applied voltage, which may depend on the conductivity and pH of the solution.

Effect of the initial pH and conductivity

Figure 6 represents the result of changing the solution's initial pH, which confirmed that conductivity is a chief parameter in achieving an effectual EC performance. It is clear that the pH would change the conductivity of the solution as well as the electrode dissociation. However, it was found that the pH varied continuously during the EC procedure [Al-Rubaiey and Albrazanjy,

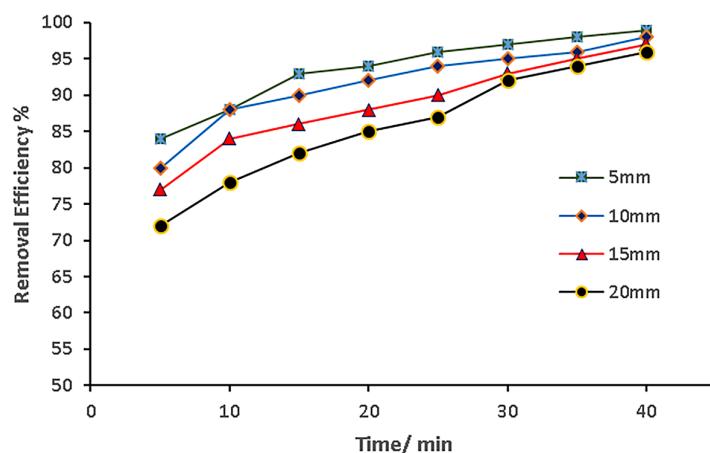


Figure 5. Effect of the electrodes' distance on the RE% of the EC wastewater treatment

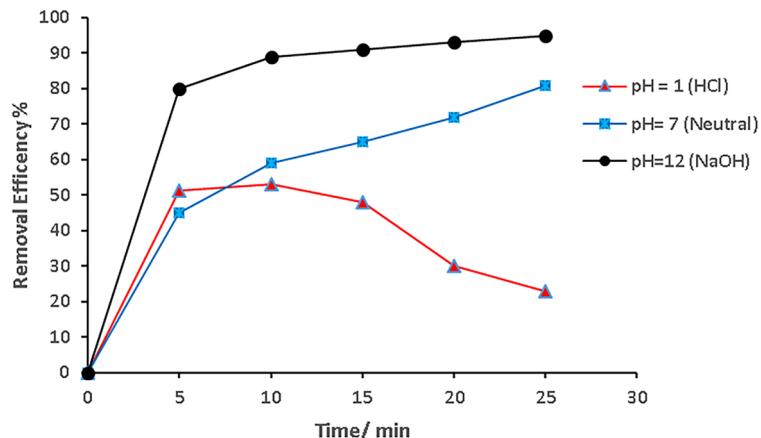


Figure 6. Effect of the initial pH on the RE% of the EC wastewater treatment

2017]. Hence, determining a linear correlation between the pH and the RE% was difficult. Thence, it was necessary to compute the starting pH to optimally influence the resultant pH in the EC process. Typically, the pH of the waste solution needed for an EC system is regulated by adding known bases, such as sodium hydroxide, or acids, such as diluted hydrochloric acid. Changing the solution pH improves the EC system because each EC procedure is related uniquely to the specific nature of the wastewater, which requires a particular ideal starting pH. For example, some wastewater provides optimal EC efficacy at acidic values, whereas some may require a relatively more alkaline pH environment [Benazzi et al., 2015]. Additionally, there is little in the literature showing that the best impurity RE% occurs with a neutral starting pH of 7. Consequently, EC is an adaptable technology whose optimal operating conditions use a wide range of pH values, mainly subject to the electrode type and their reactions with the treated pollutants [Benazzi et al., 2015; Xu and Zhu, 2004; AlJaberi et al., 2020;-Fayad, 2017]. It is obvious that the rate of suspension with dissimilar wastewater will depend on the metal being used. For example, Al metal would produce Al^{3+} ions that may be oxidized to Al hydroxide ions released near the cathode. The discharge and reactions of ions vary given the properties of the waste, necessitating a particular starting pH level for the solution. In contrast, with Fe metal, the anode produces Fe^{2+} ions, which tend to be oxidized to Fe^{3+} ions prior to the formation of iron hydroxides. Thus, the starting pH of Fe engaged in an EC process differs from that of the same process using Al metal. In addition, the pH mainly regulates the wastewater conductivity,

thus adjusting the voltage passing through the electrolyte liquid.

The pH also controls the cost viability of the electrochemical system, mainly in relation to energy consumption. Greater current conductivity leads to less current needed to attain the same impurity RE%. Furthermore, common salts (e.g., sodium chloride) have been added to increase the solution conductivity [AlJaberi et al., 2020]. This is distinctly presented in Figure 7, which demonstrates the benefits of such additions. Nevertheless, further studies are needed to enhance the electrolyte concentration for the best EC efficiency.

Effect of mixing

Figure 8 compares using an agitating mixer with a magnetic stirrer and with the procedure carried out without mixing (the mixer was turned off) under fixed conditions of 500 NTU of initial Turbidity, pH of 8, applied voltage of 10 V, electrodes distance of 0.6 cm. The results demonstrated a noticeable decrease in the removal efficiency without mixing. This reached about a 50% reduction at about 15 min of operating time. Mixing is vital in EC systems because the process was found to improve conductivity, and thus, the current density would also be amplified. Stirring is another crucial factor to confirm the uniformity of the EC solution and increase the RE% of the contaminant by providing movement throughout the mixing process. Generally, in this work, the mixing speed was fixed at one level for most EC runs for nearly all treatments, usually at about 300 rpm. However, previous papers that have examined the impact of mixing speeds on an EC system found that increased mixing would

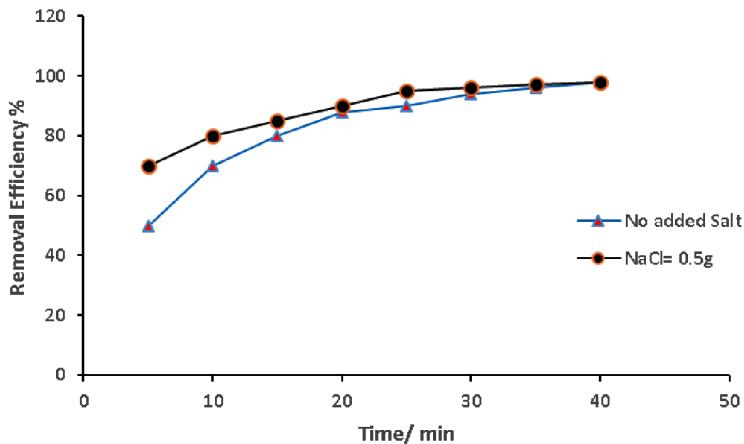


Figure 7. Effect of adding salt on the RE% of the EC wastewater treatment

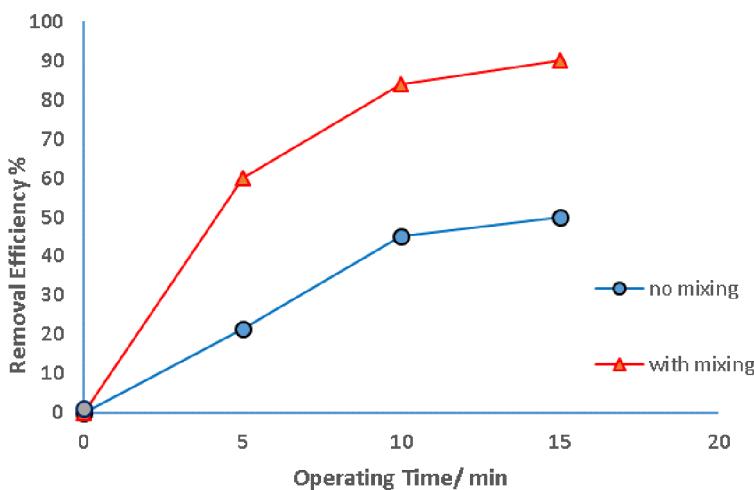


Figure 8. Effect of mixing on the RE% of the EC wastewater treatment

boost the passing of metal ions and the shaped flocs inside the solution, causing a higher RE% of the impurities [Al-Rubaiey et al., 2022]. Yet, outside the appropriate range, mixing may damage the electrocoagulation RE% by possibly re-stabilizing the colloids and thereby abolishing the formed flocs that are necessary to remove the contaminants [Fayad, 2017].

This effect was also recognized clearly in the part of our work that used an ultrasonic (US) bath partially as a mixing tool combined with the EC system. Figure 9 illustrates the impact of using US irradiation on the RE% of oily refinery wastewater. US irradiation is anticipated to improve both the kinetics and the general operations of EC remediation procedures [A-Rubaiey and Al-Barazanjy, 2018b]. Also, US irradiation would greatly improve the RE% of the EC system because the process may reduce the passivation grown on the electrodes by eliminating the solid

layer and reducing the thickness of the electrical double layer at the electrode's surface [Mahvi, 2009]. Additionally, US irradiation would activate both the ions formed in the electrode's reaction zone and the electrodes themselves by generating flaws on their surfaces. In this case, US irradiation may create friction between the liquid and the electrode surfaces, leading to local amplification. Conversely, US irradiation may also produce some negative results, including the likely annihilation of both the resulting colloidal hydroxides and the formed adsorption layer at the colloidal particles' surface [Hassani et al., 2022; Asgharian et al., 2017; Sister and Kirshankova, 2005; He et al., 2016].

Another key parameter was employing US irradiation with an EC system relates to the effect of using different types of wastewater as illustrated in Figure 10. It shows that the RE% has higher values when treating heavy metal ions in comparison

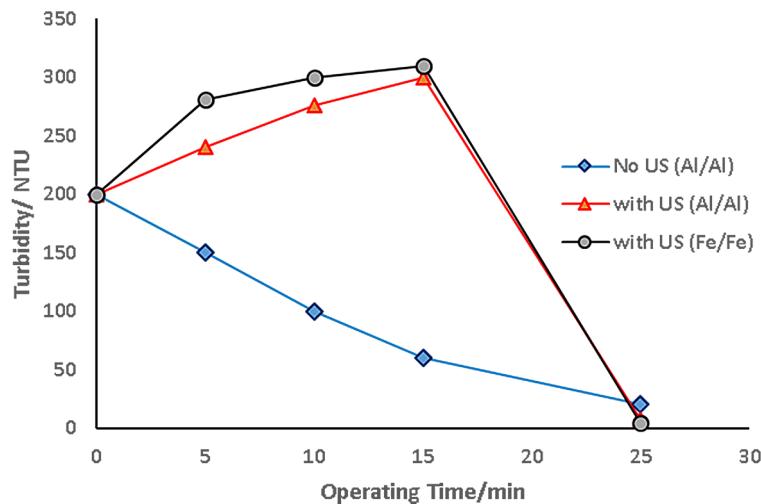


Figure 9. Effect of US irradiation and mixing on the RE% of the EC wastewater treatment

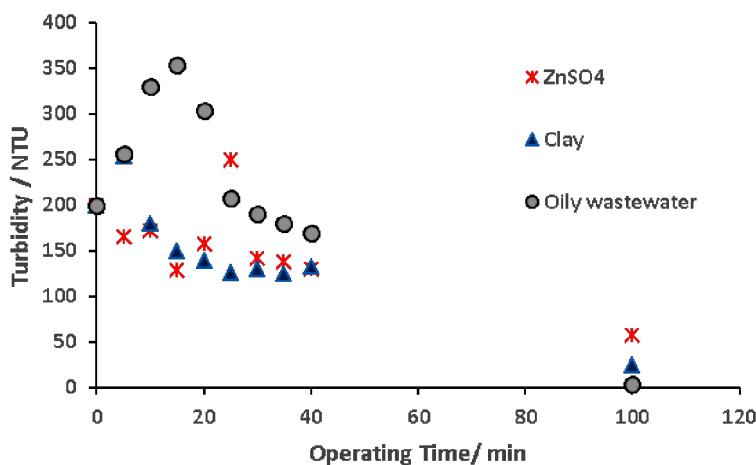


Figure 10. RE% of the EC for different wastewater effluents

to that of treating clay and oily wastewater, especially at a shorter operating time. However, further research is required to elucidate this point.

Effect of increasing the temperature

Most of the EC investigations have been conducted at ambient temperature [Sen et al., 2019]. However, studying the effect of using different temperatures on the pollutant RE% in EC procedures has mainly determined that high temperature solutions result in further operative and cost-effective colloidal removal [Jovanović et al., 2021]. This is explained because higher temperatures may result in the production of more aluminum hydroxide coagulant ions. For example, increasing the temperature from 298 K to 332 K as shown in Figure 11 considerably improved the RE% from the colloidal calcareo-argillaceous

postponement, and these higher temperatures have also been found to reduce energy consumption [Jovanović et al., 2021]. Corresponding to an increase in the RE%, running EC at high temperatures reduces the applied voltage condition, so it may effectively lower the system's costs. With higher temperatures, the foundation of extra Al hydroxides was clear because the higher rate of ion passing could be explained by Brownian motion and by collisions that could destabilize the colloids promptly with further efficacy. This also would have negatively affected the EC procedure. For instance, the coagulants tend to be more soluble at high temperatures, especially for oily wastewater; thus, at higher temperatures, the capability to detach the filterable precipitates is possibly diminished, causing ingredients to dissolve in the remediated wastewater [Sen et al., 2019; Jovanović et al., 2021].

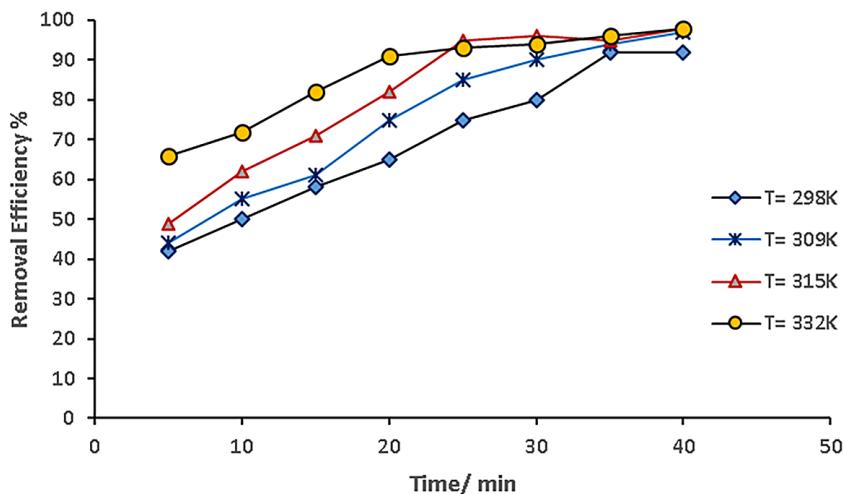


Figure 11. Effect of temperature on the RE% of the EC wastewater treatment

Table 1. Pollutants present in oily wastewater after treatment

Pollutants (ppm)	Crude wastewater	After EC treatment	After EC-PC (TiO_2) treatment
T/K	309	308	310
pH	9	7.5	7.1
Suspended solids	650	< 100	95
Oil	1900	< 50	21
BOD	< 158	< 110	< 100
COD	510	< 190	< 111
Phenols	< 2.1	< 0.6	< 0.1
Sulfides	< 61	< 2.1	< 0.5
Turbidity / NTU	500	20	10

Photocatalytic effect

One of the latest outstanding developments in EC technology for remediating several wastewater arrangements was performed with the integration of advanced oxidation processes (i.e. photocatalysis, PC) to eliminate other unremoved organic impurities [Akyol et al., 2015]. Also, the need to find a sustainable energy source (mainly for remote regions) has stimulated the exploration of employing photocatalytic EC to enhance the RE% of impurities from oily refinery wastewater as shown in Table 1. This step would encourage the use of solar energy as the main source of power since it is easily available in the Iraqi environment. The main objective of employing the combined process for industrial wastewater treatment is the use of an EC process to coagulate the large organic molecules and dye by the floccules of the metal hydroxides, thereby achieving more COD removal in a shorter time using the PC process for the remaining organic

molecules and by-products. Table 1 shows that when EC and PC were performed alone at optimum conditions, the COD decreased from 510 to < 190 mg/L (60%) and from < 190 to < 111 mg/L (40%), respectively. However, the overall COD reduction for the combined electrocoagulation photocatalytic (EC/PC) degradation process was 80%. The EC combined oxidation procedure attained a RE% of the dye of 99%, turbidity of 98%, and TDS of 85% from this industrial wastewater. Consequently, it is evident that EC/PC is a capable substitute for improving remediation efficiency while effectively reducing power costs if combined with renewable energy [Akyol et al., 2015; Gadad et al., 2016; Qing et al., 2016; Ghaffarian Khorram and Fallah, 2020; Maher et al., 2020]. The proposed mechanism for organic compound oxidation with the associated oxygen advancement occurred on the surface of the electrode through the formation of hydroxyl radicals [Al-Rubaiey and Albrazanjy, 2018].

CONCLUSIONS

Recent progress on the optimal characteristics of an EC system used for industrial wastewater remediation was reviewed, showing the growing significance and advantages of the EC procedure. The most critical features that make EC useful are its flexibility in handling a variety of industrial wastewater types, its straightforwardness, its cost efficacy, and especially, its eco-friendly sustainability when treating harmful materials. However, two considerations when employing the electrochemical system are that (1) its working parameters change depending on the type of water solution and (2) ideal operational conditions must be used. Additionally, this is an eco-friendly, reusable method of safe water recovery from treated wastewater. Using EC improved by combining it with advanced oxidation processes creates a more sustainable system than earlier, more traditional remediation methods. In addition, substituting renewable power for high-cost conventional power is a noteworthy development in using EC. Still, more work is required in terms of optimization, system design, and cost feasibility to further explore the likelihood of using hybrid EC/PC and scaling it up for use in the industrial sector.

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REFERENCES

- Abuhasel, K., Kchaou, M., Alquraish, M., Munusamy, Y., Jeng, Y.T. 2021. Oily wastewater treatment: overview of conventional and modern methods, challenges, and future opportunities. *Water*, 13, 980.
- Akyol, A., Can, O.T., Bayramoglu, M. 2015. Treatment of hydroquinone by photochemical oxidation and electrocoagulation combined process. *Journal of Water Process Engineering*, 8, 45–54.
- Alardhi, S.M., Al-Jadir, T., Hasan, A., Jaber, A., Al Saedi, L., 2023. Design of artificial neural network for prediction of hydrogen sulfide and carbon dioxide concentrations in a natural gas sweetening plant. *Ecol. Eng. Environ. Technol.*, 24(2), 55–66.
- Al Jaber, F.Y., Ahmed, S.A., Makki, H.F. 2020. Electrocoagulation treatment of high saline oily wastewater: evaluation and optimization. *Heliyon*, 6(6).
- Al-Jadir, T., Alardhi S.M., Alheety, M., Najim, A., Salih, I., Al-Furaiji, M., Alsalhy, Q. 2022. Fabrication and characterization of polyphenylsulfone/titanium oxide nanocomposite membranes for oily wastewater treatment. *J. Ecol. Eng.* 23(12): 1–13.
- Al-Rubaiey, N.A. 2022. A trends in ozone treatment of wastewater: A review. *Iraqi Journal of Oil and Gas Research*, 2(1), 55–64.
- Al-Rubaiey, N.A., Al-Barazanjy, M.G. 2017. Study the efficiency of electrocoagulation system using conductivity measurements for the removal of zinc heavy metal. In: International Conference on Environmental Impacts of the Oil and Gas Industries: Kurdistan Region of Iraq as a Case Study, 42–47.
- Al-Rubaiey, N.A., Al-Barazanjy, M.G. 2018a. Electrocoagulation treatment of oily wastewater in the oil industry. *Journal of Petroleum Research and Studies*, 8(3), 274–289.
- Al-Rubaiey, N.A., Al-Barazanjy, M.G. 2018b. Ultrasonic technique in treating wastewater by electrocoagulation. *Engineering and Technology Journal*, 36(1), 54–62.
- Al-Rubaiey, N.A., AlBarazanjy, M.G. 2019. The effect of some variables on the removal of synthetic bentonite suspension in water by electrocoagulation using turbidity measurements. In: IOP Conference Series: Materials Science and Engineering. IOP Publishing, 579(1), 012009.
- Al-Rubaiey, N.A., AlBrazanjy, M.G., Abdulkareem, M. 2020. Electrocoagulation treatment of oil-based mud wastewater. In: IOP Conference Series: Materials Science and Engineering. IOP Publishing, 737(1), 012191.
- Al-Rubaiey, N.A., AlBrazanjy, M.G., Kadhim, W.A. 2022. Combined electrocoagulation and photocatalytic for oily wastewater treatment using TiO₂ nano-catalysts. *Egyptian Journal of Chemistry*, 65(7), 55–64.
- Asfaha, Y.G., Zewge, F., Yohannes, T., Kebede, S. 2022. Investigation of cotton textile industry wastewater treatment with electrocoagulation process: performance, mineralization, and kinetic study. *Water Science and Technology*, 85(5), 1549–1567.
- Asgharian, F., Khosravi-Nikou, M.R., Anvaripour, B., Danaee, I. 2017. Electrocoagulation and ultrasonic removal of humic acid from wastewater. *Environmental Progress & Sustainable Energy*, 36(3), 822–829.
- Aswathy, P., Gandhimathi, R., Ramesh, S.T., Nidheesh, P.V. 2016. Removal of organics from bilge water by batch electrocoagulation process. *Separation and Purification Technology*, 159, 108–115.
- Bagastyo, A.Y., Sidik, F., Anggrainy, A.D., Lin, J.L., Nurhayati, E. 2022. The performance of electrocoagulation process in removing organic and

- nitrogenous compounds from landfill leachate in a three-compartment reactor. *Journal of Ecological Engineering*, 23(2), 235–245.
17. Benazzi, T.L., Dallago, R.M., Steffens, J., Bopsin, M.H. 2015. Effect of pH and conductivity in the electrocoagulation treatment of synthetic dairy effluent. *Revista Ciências Exatas e Naturais (Impresso)*, 17, 105–118.
 18. Chafi, M., Gourich, B., Essadki, A.H., Vial, C., Fabregat, A. 2011. Comparison of electrocoagulation using iron and aluminium electrodes with chemical coagulation for the removal of a highly soluble acid dye. *Desalination*, 281, 285–292.
 19. Changmai, M., Pasawan, M., Purkait, M.K. 2019. Treatment of oily wastewater from drilling site using electrocoagulation followed by microfiltration. *Separation and Purification Technology*, 210, 463–472.
 20. Chen, G., Hung, Y.T. 2007. Electrochemical wastewater treatment processes. In: *Advanced Physico-chemical Treatment Technologies*. Totowa, NJ: Humana Press, 57–106.
 21. Chen, X., Ren, P., Li, T., Trembley, J.P., Liu, X. 2018. Zinc removal from model wastewater by electrocoagulation: Processing, kinetics and mechanism. *Chemical Engineering Journal*, 349, 358–367.
 22. Demirbas, E., Koby, M. 2017. Operating cost and treatment of metalworking fluid wastewater by chemical coagulation and electrocoagulation processes. *Process Safety and Environmental Protection*, 105, 79–90.
 23. Elazzouzi, M., Haboubi, K., Elyoubi, M.S. 2017. Electrocoagulation flocculation as a low-cost process for pollutants removal from urban wastewater. *Chemical Engineering Research and Design*, 117, 614–626.
 24. Fayad, N. 2017. The application of electrocoagulation process for wastewater treatment and for the separation and purification of biological media (Doctoral dissertation), Université Clermont Auvergne, 2017–2020.
 25. Gadad, V.F., Manjunatha, B.M., Lokeshappa, B. 2016. Colour and COD removal of distillery spent wash by electrocoagulation followed by photocatalytic method. *International Journal of Advance Research in Engineering, Science & Technology*, 3(8), 30–33.
 26. Ghaffarian Khorram, A., Fallah, N. 2020. Comparison of electrocoagulation and photocatalytic process for treatment of industrial dyeing wastewater: Energy consumption analysis. *Environmental Progress & Sustainable Energy*, 39(1), 13288.
 27. Hassani, A., Malhotra, M., Karim, A.V., Krishnan, S., Nidheesh, P.V. 2022. Recent progress on ultrasound-assisted electrochemical processes: A review on mechanism, reactor strategies, and applications for wastewater treatment. *Environmental Research*, 205, 112463.
 28. He, C.C., Hu, C.Y., Lo, S.L. 2016. Evaluation of sono-electrocoagulation for the removal of Reactive Blue 19 passive film removed by ultrasound. *Separation and Purification Technology*, 165, 107–113.
 29. Jabbar, N., Alardhi S.M., Al-Jadir, T., Dhahad H. 2023. Contaminants removal from real refinery wastewater associated with energy generation in microbial fuel cell. *J. Ecol. Eng.*, 24(1), 107–114.
 30. Jovanović, T., Velinov, N., Petrović, M., Najdanović, S., Bojić, D., Radović, M., Bojić, A. 2021. Mechanism of the electrocoagulation process and its application for treatment of wastewater: A review. *Advanced Technologies*, 10(1), 63–72.
 31. Khaled, B., Wided, B., Béchir, H., Elimame, E., Mouna, L., Zied, T. 2019. Investigation of electrocoagulation reactor design parameters effect on the removal of cadmium from synthetic and phosphate industrial wastewater. *Arabian Journal of Chemistry*, 12(8), 1848–1859.
 32. Kobya, M., Ozyonar, F.U.A.T., Demirbas, E., Sik, E., Oncel, M.S. 2015. Arsenic removal from groundwater of Sivas-Şarkışla Plain, Turkey by electrocoagulation process: comparing with iron plate and ball electrodes. *Journal of Environmental Chemical Engineering*, 3(2), 1096–1106.
 33. Linares-Hernández, I., Barrera-Díaz, C., Roa-Morales, G., Bilyeu, B., Ureña-Núñez, F. 2009. Influence of the anodic material on electrocoagulation performance. *Chemical Engineering Journal*, 148(1), 97–105.
 34. Magnisali, E., Yan, Q., Vayenas, D.V. 2022. Electrocoagulation as a revived wastewater treatment method-practical approaches: a review. *Journal of Chemical Technology & Biotechnology*, 97(1), 9–25.
 35. Maher, E.K., O’Malley, K.N., Dollhopf, M.E., Mayer, B.K., McNamara, P.J. 2020. Removal of estrogenic compounds from water via energy efficient sequential electrocoagulation-electrooxidation. *Environmental Engineering Science*, 37(2), 99–108.
 36. Mahvi, A.H. 2009. Application of ultrasonic technology for water and wastewater treatment. *Iranian Journal of Public Health*, 38(2), 1–17.
 37. Mengistu, L.R., Samuel, Z.A., Kitila, C.D., Bayu, A.B. 2022. Comparison study on sonodirect and sonoalternate current electrocoagulation process for domestic wastewater treatment. *International Journal of Analytical Chemistry*, 2022.
 38. Muttaqin, R., Ratnawati, R., Slamet, S. 2022. Batch electrocoagulation system using aluminum and stainless steel 316 plates for hospital wastewater treatment. In *IOP Conference Series: Earth and Environmental Science*. IOP Publishing, 963(1), 012056.
 39. Naje, A.S., Chelliapan, S., Zakaria, Z., Ajeel, M.A., Alaba, P.A. 2017. A review of electrocoagulation technology for the treatment of textile wastewater. *Reviews in Chemical Engineering*, 33(3), 263–292.

40. Naje, A.S., Chelliapan, S., Zakaria, Z., Ajeel, M.A., Sopian, K., Hasan, H.A. 2016. Electrocoagulation by solar energy feed for textile wastewater treatment including mechanism and hydrogen production using a novel reactor design with a rotating anode. *RSC Advances*, 6(12), 10192–10204.
41. Ngala, S., Lekhlif, B., Jamal, J.E., Lakhdar, M., Afarine, L. 2019. Investigation of electrocoagulation on the removal of nickel in waste water from an electroplating bath using aluminium and iron electrodes. *Moroccan Journal of Chemistry*, 7(4), 7–41.
42. Qing, Y., Hang, Y., Xuelei, L., Hui, W., Shu, X. 2016. Combined electrocoagulation, electrolysis and photocatalysis technologies applied to ship sewage treatment. *International Journal of Environmental Science and Development*, 7(4), 248.
43. Rakhmania, Kamyab, H., Yuzir, M.A., Abdullah, N., Quan, L.M., Riyadi, F.A., Marzouki, R. 2022. Recent applications of the electrocoagulation process on agro-based industrial wastewater: a review. *Sustainability*, 14(4), 1985.
44. Rookesh, T., Samaei, M.R., Yousefinejad, S., Hashemi, H., Derakhshan, Z., Abbasi, F., Bilal, M. 2022. Investigating the electrocoagulation treatment of landfill leachate by iron/graphite electrodes: process parameters and efficacy assessment. *Water*, 14(2), 205.
45. Sathya, K., Nagarajan, K., Carlin Geor Malar, G., Rajalakshmi, S., Raja Lakshmi, P. 2022. A comprehensive review on comparison among effluent treatment methods and modern methods of treatment of industrial wastewater effluent from different sources. *Applied Water Science*, 12(4), 70.
46. Vymazal, J. 2022. The historical development of constructed wetlands for wastewater treatment. *Land*, 11(2), 174.
47. Sen, S., Prajapati, A.K., Bannatwala, A., Pal, D. 2019. Electrocoagulation treatment of industrial wastewater including textile dyeing effluent – a review. *Desalination and Water Treatment*, 161(1), 21–34.
48. Setyawati, H., Galuh, D., Yunita, E. 2021. Effect of electrode distance and voltage on Cr, COD, and TSS reduction in waste water tanning industry using electrocoagulator batch. *Journal of Sustainable Technology and Applied Sciences*, 2(1), 24–30.
49. Sharma, G., Choi, J., Shon, H.K., Phuntsho, S. 2011. Solar-powered electrocoagulation system for water and wastewater treatment. *Desalination and water treatment*, 32(1–3), 381–388.
50. Sister, V.G., Kirshankova, E.V. 2005. Ultrasonic techniques in removing surfactants from effluents by electrocoagulation. *Chemical and Petroleum Engineering*, 41(9–10), 553–556.
51. Sun, P., Elgowainy, A., Wang, M., Han, J., Henderson, R.J. 2018. Estimation of US refinery water consumption and allocation to refinery products. *Fuel*, 221, 542–557.
52. Tahreen, A., Jami, M.S., Ali, F. 2020. Role of electrocoagulation in wastewater treatment: A developmental review. *Journal of Water Process Engineering*, 37, 101440.
53. Um, S.E., Chang, I.S. 2017. Effect of current density and contact time on membrane fouling in electrocoagulation-MBR and their kinetic studies on fouling reduction rate. *Journal of Korean Society of Water and Wastewater*, 31(4), 321–328.
54. UNEP in Iraq: Post-Conflict Assessment, Clean-up and Reconstruction. Report 2007, United Nations Environment Programme. Job No.: DEP/1035/GE.
55. Xu, X., Zhu, X. 2004. Treatment of refractory oily wastewater by electro-coagulation process. *Chemosphere*, 56(10), 889–894.
56. Zeboudji, B., Drouiche, N., Lounici, H., Mameri, N., Ghaffour, N. 2013. The influence of parameters affecting boron removal by electrocoagulation process. *Separation Science and Technology*, 48(8), 1280–1288.